Coexistence of ferromagnetism and Kondo effect in uranium compounds

N. B. Perkins,[1,2] J. R. Iglesias,[3] M. D. Núñez-Regueiro,[4] and B. Coqblin[4]

[1]MPIPKS, Nöthnitzer Str. 38, 01187 Dresden, Germany
[2]Bogoliubov Laboratory of Theoretical Physics, JINR, Dubna, Russia
[3]Instituto de Física, Universidade Federal do Rio Grande do Sul, 91501-970 Porto Alegre, Brazil
[4]Laboratoire de Physique des Solides, Université Paris-Sud, Bâtiment 510, 91405 Orsay, France

Coexistence between ferromagnetic order and Kondo behavior has been observed in some uranium compounds. The underscreened Kondo lattice model can provide a possible description of this coexistence. Here we present a model of a lattice of $S = 1$ spins coupled to the conduction electrons through an intra-site exchange interaction J_K and an inter-site ferromagnetic exchange $f-f$ interaction J_H. Finite temperature results show that the Kondo temperature is larger than the Curie ordering temperature, T_C, providing a possible scenario for the coexistence of Kondo effect and magnetic order. Also, the Kondo behavior disappears abruptly for low values of J_K and smoothly when changing the band occupation. These results are in qualitative agreement with the experimental situation for the above mentioned uranium compounds.

PACS numbers: 71.27.+a, 75.30.Mb, 75.20.Hr, 75.10.-b

Many rare-earth and actinide compounds show anomalous properties which have been attributed to heavy fermion behavior. A lot of attention has been placed on the experimental and theoretical study of the competition between magnetic order - mainly antiferromagnetic (AF) - and Kondo effect in cerium or ytterbium compounds [1, 2]. As a result, nowadays there exist various available theories describing many reliable experiments, or vice versa. For a review see ref. [1].

On the other hand, uranium compounds present many distintic behaviors from mixed valence to magnetic order with or without a Kondo effect and even a coexistence between ferromagnetic order and superconductivity, as it has been observed in UGe_2 and $URhGe$ [3].

However, one of the most interesting experimental findings, the coexistence of Kondo behavior and ferromagnetic order observed in some uranium compounds has been – to our knowledge – somewhat overlooked from a theoretical point of view. It's a long way since the first experimental evidence of coexistence between Kondo behavior and ferromagnetic order in the dense Kondo compound UTe [4]. More recently this coexistence has been observed in $UCu_{0.9}Sb_2$ [5] and $UCo_{0.5}Sb_2$ [6]. Those systems undergo a ferromagnetic ordering at relatively high Curie temperatures of T_c = 102K, T_c = 113K and T_c = 64.5K, correspondingly, and also present a logarithmic decrease of the magnetic resistivity above it, suggesting a Kondo behavior at high temperature. These experimental results clearly show the coexistence of ferromagnetic order and Kondo behavior below a large Curie temperature. Another example of such coexistence has been found in $URu_{2-x}Re_xSi_2$ compounds, where a non-Fermi liquid (NFL) behavior of resistivity and specific heat has been observed inside the ferromagnetic phase [7, 8].

In order to account for the Kondo-ferromagnetism coexistence observed in the above quoted uranium compounds, we propose the framework of the underscreened Kondo lattice (UKL) model, that incorporates all the essential aspects of the problem such as Kondo interaction and magnetic RKKY interaction between localized spins $S = 1$. We believe it is reasonable to assume a $5f^2$ configuration for uranium because the magnetic moments deduced from magnetic experiments in these compounds are close to the free ion values [9] and moreover a good agreement with experiments has been obtained by considering only 2 localized 5f electrons [10].

The underscreened Kondo impurity model has been studied using different approaches[11] and an exact solution was obtained by the Bethe-ansatz method [12]. On the other hand there are relatively few studies of the UKL, among them refs.[13, 14]. Here we describe the UKL as a periodic lattice of magnetic atoms with $S = 1$ – which can be modeled by two degenerate f-orbitals – interacting with itinerant spins. The spins of these f-electrons are coupled to $S = 1$ due to the strong on-site Hund's coupling and we treat them on the basis of a fermionic representation for spin $S = 1$ [15], valid in this constrained triplet Hilbert space. Then, we perform a mean field analysis of the model, both at zero- and finite-temperatures in terms of the Green's functions of the system.

In normal Kondo lattice (KL) model ($S = 1/2$, i.e. one f-electron per lattice site) the competition between Kondo effect and magnetic RKKY interaction leads to the appearance of a quantum phase transition between a magnetically ordered (generally antiferromagnetic) phase and a phase with coherent Kondo spin-singlet formation and short range magnetic correlations [2, 16]. In a similar way, with the help of the UKL model we can describe quantum ferromagnetic phase transitions and the coexistence between ferromagnetic order and NFL behavior [8] at different values of external parameters such as band filling, pressure or temperature. Based on this analysis we can draw a phase diagram showing magnetic ordered phases with no Kondo effect and regions of coexistence between ferromagnetic order and Kondo behavior.

The UKL Hamiltonian can be written in the following form:

$$H = \sum_{\vec{k},\sigma} (\epsilon_{\vec{k}} - \mu) \mathbf{n}^c_{\vec{k}\sigma} + \sum_{i\sigma\alpha} E_0 \mathbf{n}^{f\alpha}_{i\sigma} +$$

$$J_K \sum_i \mathbf{S}_i \sigma_{\mathbf{i}} + \frac{1}{2} J_H \sum_{ij} \mathbf{S}_i \mathbf{S}_j \tag{1}$$

where the first term represents the conduction band with dispersion ϵ_k, width $2D$ and a constant density of states $1/2D$, while μ is the bare electron chemical potential. Localized spins are represented by fermionic operators $\mathbf{f}^{\dagger}_{\mathbf{i}\sigma\alpha}$ and $\mathbf{f}_{\mathbf{i}\sigma\alpha}$ introduced in refs.[13, 15], carrying spin, σ, and orbital, α, indexes. E_0 is a Lagrange multiplier which is fixed by a constraint for the total number of f-electrons per site, $n_f = \sum_\sigma (n^{f1}_\sigma + n^{f2}_\sigma) = 2$, and can be interpreted as a fictitious chemical potential for f-fermions. The third term is the on-site Kondo coupling, $J_K > 0$, between localized $\mathbf{S_i} = 1$ and conduction electron's $\sigma_i = 1/2$ spins, whose fermionic representation is done in the usual way. And finally, the last term is the ferromagnetic inter-site interaction, $J_H < 0$, between localized f-magnetic moments, resulting explicitly from two contributions: the effective RKKY interaction, and the direct exchange.

As a first step we introduce the relevant fields. They are $\widehat{\lambda}_{i\sigma} = \sum_\alpha \mathbf{c}^+_{\mathbf{i}\sigma} \mathbf{f}^\alpha_{\mathbf{i}\sigma}$ which couple $c_{i\sigma}$ and $f^\alpha_{i\sigma}$ electrons at the same site, and operators of magnetization for both c- and f-subsystems, which are, respectively, $\mathbf{M_i} = S^z_i = \frac{1}{2}(n^f_{i\uparrow} - n^f_{i\downarrow})$ and $\mathbf{m_i} = \sigma^z_i = \frac{1}{2}(n^c_{i\uparrow} - n^c_{i\downarrow})$ describing the long range magnetic order of the system. Then we perform a mean field (MF) analysis of the Hamiltonian eq. (1), expanding it according to the four following averages: $\lambda_\sigma = \langle \widehat{\lambda}_{i\sigma} \rangle$, $M = \langle \mathbf{M_i} \rangle$ and $m = \langle \mathbf{m_i} \rangle$. The non-zero values of M and m correspond to the formation of the magnetic phase with finite total magnetization, while the parameter λ_σ describes an effective hybridization between conduction and $f-$ electrons that corresponds to the Kondo behavior. In MF approximation the average value of $\sum_\alpha c^+_{i\sigma} f^\alpha_{i\sigma}$ may be different from zero, implying spurious charge fluctuations. But one should keep in mind that the original term of the Hamiltonian is a four-fermion operator, i.e. a product $\widehat{\lambda}_\sigma \widehat{\lambda}_{\bar{\sigma}}$ [17]. So, λ_σ should be interpreted as a tool for the MF calculation that provides a correct description of the Kondo temperature, for example in the study of the Kondo impurity [18]. This procedure is basically equivalent to other mean field approaches developed for studying normal KL, such as the path-integral calculation restricted to a saddle-point solution performed by Coleman and Andrei[19], or large-N formulation performed by Burdin et al [20], where again the saddle-point solution yields a mean-boson-field approximation.

The resulting MF Hamiltonian reads

$$\begin{aligned} H_{MF} &= \textstyle\sum_{\vec{k}\sigma} \varepsilon_{\vec{k}\sigma} n^c_{\vec{k}\sigma} + \sum_{i\alpha\sigma} E_{0\sigma} n_{i\alpha\sigma} - \\ &- \tfrac{1}{2} J_K \textstyle\sum_{i\alpha\sigma} (\lambda_{\bar{\sigma}} \widehat{\lambda}^\alpha_{i\sigma} + h.c.) + \\ &2 J_K N \textstyle\sum_\sigma \lambda_{\bar{\sigma}} \lambda_\sigma - J_K N m M - \tfrac{1}{2} J_H N z M^2 \end{aligned} \tag{2}$$

where, remembering that $J_K > 0$ and $J_H < 0$, energies read

$$\begin{aligned} \varepsilon_{\vec{k}\sigma} &= \varepsilon_{\vec{k}} + J_K \sigma M \;,\; with \quad \sigma = \pm 1/2 \\ E_{0\sigma} &= E_0 + J_K \sigma m - \tfrac{1}{2} J_K \lambda_\sigma \lambda_{\bar{\sigma}} + J_H z \sigma M \end{aligned} \tag{3}$$

The diagonalization of the MF Hamiltonian (2) gives two non-hybridized f-bands (one for each spin) at energies $E_{0\sigma}$ and two quasiparticle bands $E^\sigma_\pm(\vec{k})$ with energies

$$E^\sigma_\pm(\vec{k}) = \frac{1}{2}[E_{0\sigma} + \varepsilon_{\vec{k}\sigma} \pm \sqrt{(E_{0\sigma} - \varepsilon_{\vec{k}\sigma})^2 + 8\alpha^2_{\bar{\sigma}}}] \tag{4}$$

where $\alpha_{\bar{\sigma}} = -\frac{1}{2} J_K \lambda_{\bar{\sigma}}$. The $\pm$ sign refers to the upper (lower) hybridized band. Let us point out here one of the main differences between the mean field treatment of the normal KL and UKL models: in the underscreened case one f-localized level remains non-hybridized and for the other level the resulting $c - f$ effective hybridization is twice that of the normal case. The energy spectra $E^\sigma_\pm(k)$ depend on a set of external parameters such as band filling n_c, Kondo coupling J_K and exchange interaction J_H and a set of internal parameters, M, m, λ_σ, μ and E_0, which should be calculated in a self-consistent way. The system of self-consistent equations can be obtained by keeping constant the number of f- and c- electrons. Their expression can be evaluated from the Green functions of the system by straightforward calculations :

$$\begin{aligned} & n^\sigma_f = \tfrac{1}{2D} \textstyle\int_{-D+\Delta_\sigma}^{D+\Delta_\sigma} d\varepsilon_\sigma [n_F(E_{0\sigma}) - \\ & n_F(E_{+\sigma}) \tfrac{\varepsilon_\sigma - E_{+\sigma}}{W_\sigma(\varepsilon_\sigma)} + n_F(E_{-\sigma}) \tfrac{\varepsilon_\sigma - E_{-\sigma}}{W_\sigma(\varepsilon_\sigma)}] \\ & n^\sigma_c = \tfrac{1}{2D} \textstyle\int_{-D+\Delta_\sigma}^{D+\Delta_\sigma} d\varepsilon_\sigma [-n_F(E_{+\sigma}) \tfrac{E_{0\sigma} - E_{+\sigma}}{W_\sigma(\varepsilon_\sigma)} + \\ & n_F(E_{-\sigma}) \tfrac{E_{0\sigma} - E_{-\sigma}}{W_\sigma(\varepsilon_\sigma)}] \end{aligned} \tag{5}$$

where $n_F(\omega) = \frac{1}{e^{\frac{\omega - E_F}{T}} + 1}$ is the Fermi distribution function, $\Delta_\sigma = J_K \sigma M$ and $W_\sigma(\varepsilon) = \sqrt{(E_{0\sigma} - \varepsilon)^2 + 8\alpha^2_{\bar{\sigma}}}$. From the Green functions we can also obtain the expression for λ_σ given by:

$$\lambda_\sigma = \tfrac{1}{D} \textstyle\int_{-D+\Delta_\sigma}^{D+\Delta_\sigma} d\varepsilon_\sigma [n_F(E_{+\sigma}) - n_F(E_{-\sigma})] \tfrac{\alpha_{\bar{\sigma}}}{W_\sigma(\varepsilon_\sigma)} \tag{6}$$

In order to construct a set of self-consistent equations defined by (5) and (6), we substitute the strong constraint, $n_{if} = 2$ in each site, by a softer one for its average, and set the band filling by the averaged number of c-electrons, n_c, which we consider in the usual range of partial filling, i.e. $0 < n_c < 1$:

$$n_f = n^\uparrow_f + n^\downarrow_f = 2 \;, \quad n_c = n^\uparrow_c + n^\downarrow_c \tag{7}$$

Having solved the system of self-consistent equations and also minimized the free energy, we study various properties of the UKL model. In Fig.1 we present a summary of the results for $T = 0$. The region of the parameters J_K and n_c that provide finite values of $\lambda_\uparrow$ corresponds to a coexistence between the heavy fermion behavior and ferromagnetic order. The values of $\lambda_\downarrow$ are close to $\lambda_\uparrow$ so the former is not depicted on the figure, neither the magnetization, to preserve clearness. It is possible to see on Fig.1 that $\lambda_\uparrow$ decreases smoothly as a function of n_c, following an approximate square root behavior [2], while it undergoes a sharp transition as a function of J_K. When $\lambda_\uparrow$ goes to zero the ground state is magnetically ordered with no Kondo effect. The hybridization gap $\Gamma_\sigma = 8\alpha^2_{\bar{\sigma}}$

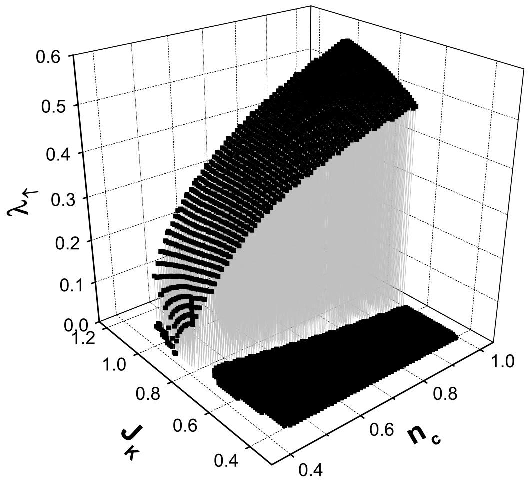

FIG. 1: Plot of the parameter $\lambda_\uparrow$ as a function of J_K and n_c for $T = 0$. The ferromagnetic interaction between localized f-moments is $J_H = -0.01$. There is a discontinuous transition as a function of J_K and a behavior in $\sqrt{n_c}$ as explained in the text

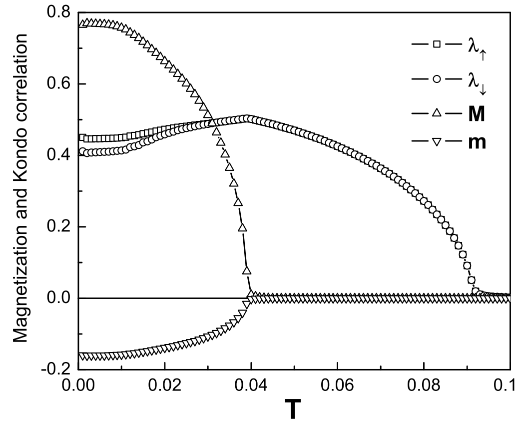

FIG. 2: Plot of $\lambda_\uparrow$, $\lambda_\downarrow$, M and m as a function of temperature. Self-consistent solutions are obtained for the following parameters:$J_K = 0.8$, $J_H = -0.01$ and $n_c = 0.8$ (in units of the half-bandwidth D).

vanish simultaneously, signaling a quantum phase transition. Detailed calculations and results including a full phase diagram with different values of J_K and J_H will be presented in a forthcoming article.

Finite temperature calculation permits to determine the Curie temperature and the correlation (Kondo) temperature. Fig.2 shows the behavior of the $f-$ and $c-$ magnetizations, M and m respectively, and λ_σ, as a function of the temperature. The parameters here and in following figures are $J_K = 0.8$, $J_H = -0.01$ and $n_c = 0.8$ and all energies and temperatures are in units of the half-bandwidth D. At zero and low temperatures we observe the coexistence of magnetic order and heavy fermion behavior, but as both spin systems are strongly polarized the Kondo effect is concealed. When the magnetization decreases, λ_σ grows, having its maximum at the Curie temperature when the magnetization vanishes. Due to the breakdown of the spin symmetry $\lambda_\downarrow$ and $\lambda_\uparrow$ are slightly different in the magnetic region but they coincide when the magnetization vanishes for $T = T_C$, i.e. when the spin symmetry is restored. For $T > T_C$ the system exhibits only Kondo behavior ($\lambda_\sigma \neq 0$, $M = 0$ and $m = 0$). Finally, above a characteristic temperature, the Kondo temperature, T_K, both λ_σ vanish and the two electron systems are decoupled.

This behavior can be further clarified by the plot of the densities of states at different temperatures. As the $c-$densities of states are almost constant (except in the hybridization gap region) we present the results only for the $f-$densities of states, which are calculated numerically from the imaginary part of the $f - f$ Green functions.In Fig.[3] we plot $\rho_{f\sigma}(\epsilon - E_F)$ for four different temperatures. At zero and low temperatures, in the magnetic phase, $T < T_C$, the Fermi level lies inside the hybridization gap for the up-band, but inside the conduction band for down-spin, $E_-^\downarrow(k)$, which implies a semi-metal behavior in the magnetic phase. Then, an insulating phase is obtained for $T > T_C$ when the Fermi level is inside the gap for both up and down spin directions and coincides with the energy of the f-level, E_0, (see, Fig.[3c and d]). Finally, when λ_σ goes to zero, the hybridization gap closes and the system becomes metallic, but with a strong heavy fermion behavior due to the fact that the Fermi level coincides with the localized f-level, E_0.

At last, from the quasiparticle spectrum (4) one can estimate the mass enhancement, which will be also spin-dependent [21] :

$$\frac{m^*_\sigma}{m} = 1 + \frac{2\alpha^2_{\bar{\sigma}}}{(E_{0\sigma} - E_F - \Delta_\sigma)^2} \tag{8}$$

We can see in the Fig.4 that for both spin directions the mass enhancement increases with temperature and becomes dramatic in the pure Kondo regime, when the magnetization goes to zero. As the denominator of (8) goes to zero in the pure Kondo region the effective mass goes strictly to infinite. But in order to better visualize its behavior and particularly the second transition at T_K, we included a very small finite width in the $f-$level so obtaining extremely high values for the effective mass in the pure Kondo region. Hence, two peaks are evident, one corresponds to the Curie temperature and the second, at higher temperatures, to the Kondo temperature. In the region of coexistence, the effective mass increases as a function of temperature following the position of the Fermi level inside the $E_-^{\bar{\sigma}}(k)$ band. Although simplified, this result provides a qualitatively good explanation of the behavior of the electronic specific heat.

In conclusion, we have formulated a mean field theory of coexistence of ferromagnetism and heavy fermion behavior in terms of self-consistent equations for the relevant fields. Within this treatment of the UKL model we obtain - for values of J_K between 0.7 and 1.2 - a region where the order parameters, λ_σ, the $f-$magnetization, M, and the $c-$magnetization, m, are different from zero, characterizing a coexistence between the heavy fermion (Kondo) properties and ferromag-

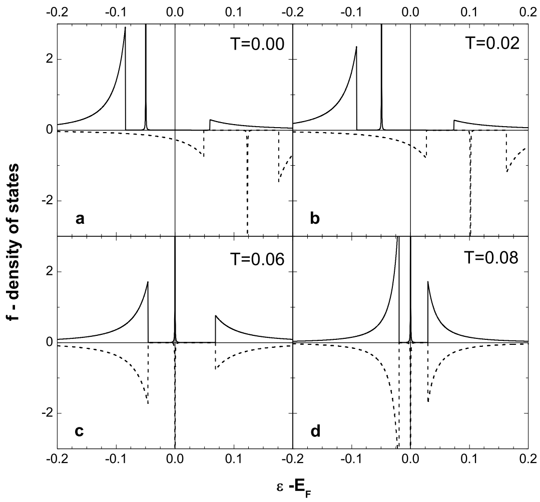

FIG. 3: Temperature variation of the up- (solid line) and down- (dashed line) f-density of states, $\rho_{f\sigma}(\epsilon - E_F)$. Panel (a) corresponds to $T = 0$, (b) $T = 0.02$, (c) $T = 0.06$ and (d) $T = 0.08$. The parameters are the same as in Fig.2.

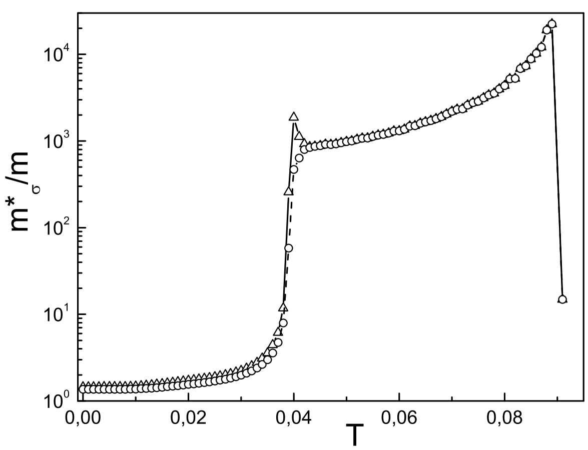

FIG. 4: Temperature dependence of the effective mass enhancement. The parameter set is the same as in Fig.2.

netic order. When increasing temperature the magnetic order disappears at the ordering temperature T_C while λ_σ is still different from zero, so indicating a Kondo or NFL regime. This is in agreement with experiments for uranium compounds where a Kondo behavior is observed also above the Curie temperature, like the examples we have quoted before. We remark also that the characteristic Kondo temperature, T_K, increases when the Kondo coupling value J_K also increases. Those results are completely different from the previous ones obtained for the normal KL model, where the Kondo effect and the magnetic order are in competition and cannot be present together [2, 16]. The critical temperature T_C characterizes here a continuous quantum phase transition from the ferromagnetic Kondo phase to the non-magnetic Kondo phase at higher temperatures. Another transition can be obtained at $T = 0$ by varying Kondo coupling parameter J_K, but this time it is discontinuous and leads to a non-Kondo magnetically ordered state, as it can be observed in Fig.1. We remark that this coexistence is mainly due to the presence of both one non-hybridized and one hybridized f-level (see Fig.3), and this ingredient is the key for the existence of the ferromagnetically ordered Kondo state. Thus, the proposed UKL model can explain the behavior of some uranium compounds where a ferromagnetically ordered state coexists with a heavy fermion Kondo state. The FM-Kondo coexistence arises from the fact that the localized spins are only partially screened, and consequently they keep their magnetic interaction at low temperatures.

The authors aknowledge stimulating discussions with S. Burdin, P.Fulde, R. Ramazashvili and J. Schoenes. JRI thanks MPIPKS, Dresden, and MPIDSO, Göttigen, for hospitality. The authors would like to acknowledge Brazilian agencies CNPq and FAPERGS for financial support.

[1] B. Coqblin, M. D. Núñez-Regueiro, Alba Theumann, J. R. Iglesias and S. G. Magalhaes, Phil. Mag. **86**, 2567 (2006) and references therein
[2] B. Coqblin, C. Lacroix, M. S. Gusmao and J. R. Iglesias, Phys. Rev. B **67**, 064417 (2003)
[3] J. Flouquet, Progress in Low Temperature Physics, **15**, 139-281, ed. by W.P. Halperin, Elsevier (2006)
[4] J. Schoenes, J. Less-Common Met., **121**, 87 (1986)
[5] Z. Bukowski, R. Troc, J. Stepien-Damm, C. Sulkowski and V. H. Tran, J. Alloys and Compounds, **403**, 65 (2005).
[6] V. H. Tran, R. Troc, Z. Bukowski, D. Badurski and C. Sulkowski, Phys. Rev. B **71**, 094428 (2005).
[7] Y. Dalichaouch, M. B. Maple, M. S. Torikachvili and A. L. Giorgi, Phys. Rev. B **39**, 2423 (1989)
[8] E. D. Bauer, V. S. Zapf, P.-C. Ho, N. P. Butch, E. J. Freeman, C. Sirvent and M. B. Maple, Phys. Rev. Lett. **94**, 046401 (2005)
[9] J. Schoenes, O. Vogt, J. Lohle, F. Hulliger, and K. Mattenberger, Phys. Rev. B **53**, 14987 (1996)
[10] G. Zwicknagl, A. Yaresko and P. Fulde, Phys. Rev. B **68**, 052508 (2003)
[11] J. Gan, P. Coleman and N. Andrei, Phys. Rev. Lett. **68**, 3476 (1992).
[12] P. Schlottmann and P. D. Sacramento, Adv. in Physics **42**, 641 (1993)
[13] N. B. Perkins, M. D. Núñez-Regueiro, J. R. Iglesias and B. Coqblin, Physica B $\mathbf{378-380}$, 698 (2006).
[14] S. Florens, Phys. Rev. B **70**, 165112 (2004)
[15] S. Di Matteo, N.B. Perkins and C.R Natoli, Phys. Rev. B **65**, 054413 (2002).
[16] J.R. Iglesias, C. Lacroix and B. Coqblin, Phys. Rev. B **56**, 11820 (1997).
[17] S. Burdin, Thesis, Grenoble (2001)
[18] C. Lacroix and M. Cyrot, Phys. Rev. B **20**, 1969 (1979).
[19] P. Coleman and N. Andrei, J. Phys.: Condens. Matter **1**, 4057 (1989).
[20] S. Burdin, A. Georges and D. R. Grempel, Phys. Rev. Lett. **85**, 1048 (2000)
[21] V. Barzykin, Phys. Rev. B **73**, 094455 (2006).